Part II

Carbon Monoxide Distributions and Atmospheric Transports over Southern Africa

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Observations of CO at Midtropospheric Levels Over the South Atlantic and Adjacent Continents

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Abstract

Sources and transports of CO as measured by the MAPS over a substantial sector of the southern hemisphere between South America and southern Africa are described by air parcel trajectories based upon ECMWF model data fields. Observations of vegetation fires made by NASA Shuttle astronauts during the October 1994 mission suggest a direct relationship between in situ biomass burning, at least over South America and southern Africa, and coincident tropospheric measurements of CO. Results of this paper indicate that the transport of CO from the surface to the levels of maximum MAPS sensitivity (~450 hPa) over these regions is not of a direct nature due largely to the well stratified atmospheric environment. The atmospheric transport of CO from biomass burning within this region is found to occur over intercontinental scales over numbers of days to more than a week. Three distinct synoptic circulation and transport classes are found to have occurred over southern Africa during the October 1994 MAPS experiment: transport from South America and Africa to southern Africa associated with elevated MAPS measured CO (>150 ppbv); weakening anticyclonic transport from South America associated with moderate CO (<150 ppbv and >105 ppbv); and transport from the high southern latitudes associated with low CO (<105 ppbv).

1. Introduction

Carbon monoxide (CO) is produced at the earth's surface and has a lifetime in the atmosphere of a few weeks to a few months (Connors et al., 1996). Biomass burning is considered to be a significant source of CO (Levine et al., 1991, 1996; Andreae, 1991). Levine et al. (1996a,b) also found that CO fluxes from the soils of southern African savannas were considerably higher than previously reported. Savanna soil fluxes of CO were observed to increase with increasing soil temperature and were strongly enhanced by burning. Combustion of fossil fuels represents the other major source of CO.

Carbon monoxide undergoes horizontal and vertical transports in the atmosphere distributing the gas vertically through the depth of the troposphere and horizontally over distances of thousands of kilometers. Global distributions of CO in the atmosphere were difficult to determine prior to 1981 when estimates of the distribution of CO in the atmosphere were based upon surface measurements at only a few locations and on limited aircraft flights. In November 1981, the Measurement of Air Pollution from Satellites (MAPS) instrument was flown on the second flight of the NASA Space Shuttle (Reichle et al., 1986). MAPS yielded the first truly global picture of the tropospheric distribution of CO. The November 1981 flight of MAPS showed maximum values of CO in the tropics between South America and Africa contrary to the canonical view of the time which held that the maximum in CO mixing ratios was in the northern hemisphere. Strong gradients of CO in latitude and longitude were shown by the 1981 MAPS flights and even more strikingly by the October 1984 flight with very high mixing ratios observed immediately "downwind" over the Atlantic and Indian Oceans from South

America and southern Africa, respectively (Connors et al., 1991; Newell et al., 1989; Reichle et al., 1990).

In 1994 a modified and improved MAPS instrument was flown on the space shuttle Endeavour on 9-19 April and 30 September-11 October (Connors et al., 1996). The April 1994 measurements showed maximum global values of CO mixing ratios in high northern latitudes with zonal gradients much weaker than meridional gradients (Figure 1). In October 1994, the global distribution was similar to those of the October 1984 flight but with significant increases in geographic areas, particularly the maritime continent, affected by elevated CO mixing ratios (Reichle and Connors, 1998). Highest CO values were located in the tropics with marked longitudinal variations showing three centers of maximum concentrations (> 135 ppbv) illustrated in Connors et al. (1998; Figure 9). The three areas of maximum concentration are the Amazon Basin and the western tropical south Atlantic Ocean, sub-equatorial Africa and the eastern tropical south Atlantic Ocean and the eastern equatorial Indian Ocean. Observations of vegetation fires made by the shuttle astronauts during the October 1994 flight (Figure 2) suggest that the observed high concentrations of CO are directly related to in situ biomass burning at least over South America and southern Africa.

This paper will explore the association between the elevated concentrations of CO as measured by MAPS during the October 1994 time period and both specific sources and source regions.

2. Methods and Data

The MAPS instrument determines total columnar CO concentrations with a nadir-viewing gas filter correlated radiometer operating at 4.67 micrometers (Reichle et al., 1986; 1998). The

instrument makes observations in the troposphere with particular emphasis on the middle and upper (~ 400 hPa) layers. The MAPS instrument is sensitive to CO between 3 and 12 km with maximum signal between 8 and 10 km or pressure levels between 450 and 400 hPa and least sensitive to CO mixing ratios below 2 km and above 12 km (Reichle et al., 1990; Pougatchev and Sachse, 1997; Reichle et al., 1998; Reichle and Connors, 1998). The CO mixing ratios as measured by MAPS during the April and October shuttle missions were found to be within 10% of those measured by aircraft flying both vertical profiles and constant level cruises at an altitude of 8-10 km (Reichle et al., 1998). At other altitudes, particularly in the surface and boundary layers, the MAPS measurement is not affected by CO enhancements near the source locations and will not show elevated CO mixing ratios there unless the CO enriched air is transported to the middle and upper troposphere.

The large-scale circulation fields of the atmosphere over tropical and subtropical southern Africa and South America are shifting in early October from the extremely dry conditions of winter to the wetter summer regime. The large semi-permanent subtropical high pressure systems responsible for the dry winter conditions produce pervasive mid-tropospheric (~ 450 hPa) inversions and stable layers (Tyson et al., 1996a; Cosijn and Tyson, 1996). Trace gases, including CO, and aerosols become trapped and concentrate immediately below this stable layer (Tyson et al., 1996b). Because of the relatively long residence time of CO in the atmosphere, concentrations of CO below the 450 hPa stable layer are pronounced. Concentrations of CO below 2.5-3km above the southern African surface, as observed during SAFARI/TRACE-A, are on the order of 300 to 400 ppbv, with values at times exceeding 600 ppbv (Blake et al. 1996).

Accumulation of surface-derived species in the troposphere over southern Africa under the anticyclonically dominated conditions of late winter and early spring is further enhanced by the circulation itself. Products injected into the atmosphere over the subcontinent are trapped within this recirculating gyre that is centered over the land mass with typical residence times within this gyre of more than two weeks (Tyson et al., 1996c). While such conditions have been explicitly demonstrated and documented over South Africa, it is likely that the anticyclone over northeastern Brazil creates a similar situation at this time of year over South America.

3.   Results

Based on the MAPS CO mixing ratios over southern Africa and the immediately adjacent ocean areas, three distribution classes can be suggested:

1)   *High*   CO mixing ratios, > 150 ppbv,

2)   *Moderate*   105 ppbv < CO mixing ratios < 150 ppbv, and

3)   *Low*   CO mixing ratios, < 105 ppbv.

Ten of the 11 days of observations by MAPS during October 1994, can be placed within one of the above three classes: Class 1 (5 days); Class 2 (3 days); and Class 3 (2 days). One day, due to orbit configuration, contains insufficient data to allow classification. Figure 3 shows representative CO maps of each of the above three classes.

The origin of backward air parcel trajectories, initiated at the 450 hPa level, was chosen at two locations on the west and east coasts of southern Africa at respectively 21° S, 12° E and 25° S, 34° E. These sites have previously been identified by Garstang et al. (1996) and Swap et al.

(1996) as close to the climatologically - dominant transport pathways present over southern Africa (Tyson et al., 1996a).

Individual air parcel trajectories can exhibit considerable spatial variability when extended over time periods as long as 10 days (D'Abreton, 1996; Pickering et al., 1996). A cluster of nine three-dimensional trajectories originating from a 2° x 2° latitude and longitude grid centered on the point of origin is calculated to provide a representative pathway potentially followed by the air (Figure 4). The degree of dispersion of the 9-trajectory cluster provides a qualitative measure of confidence when determining the source and pathway of the air. Nine point trajectories extending backwards for 10 days from the points of origin on the east and west coasts of southern Africa are presented in Figure 5 for each of the three classes shown in Figure 3.

For the high CO class, where individual cells of high values of CO mixing ratios occur over the entire region (Figure 3a), air parcels at 450 hPa on the west coast of southern Africa originate primarily from the lower troposphere over central sub-equatorial Africa (Figure 5a). On the east coast of southern Africa (Figure 5b) trajectories cross the Atlantic in a tight cluster originating both over the southern Amazon Basin and over southern Brazil and Paraguay. In both cases trajectories originate in regions where many fires are observed (Figure 2) but from altitudes above 400 hPa. Although biomass burning emissions are trapped vertically over southern Africa, vertical transport of biomass burning emissions associated with deep convection from the surface to these heights at this time of the year has been demonstrated for the Amazon basin (Pickering et al., 1996; Thompson et al., 1996). Transports over southern Africa and over the south Atlantic are a function of well - established anticyclones over both locations. Over southern Africa, the back trajectories on the west coast reflect the transports due to the continental

anticyclone with trajectories originating as far north and east as Kenya. Inflow over the southwestern Cape coast feeds into the southern arc of the continental high. The Subtropical Anticyclone (STAC) over the south Atlantic is displaced equatorward such that westerly flow embedded in the southern limb of the STAC entrains air over eastern Brazil transporting it directly across the Atlantic to South Africa.

With the example from the moderate CO class, no high values are seen over eastern southern Africa or over the south Indian Ocean (Figure 3b). Corresponding back trajectories particularly over southern Africa, have lost their strong anticyclonic curvature and suggest a decay of the anticyclone over southern Africa (Figure 5c and d). The STAC has also weakened and westerly transport across the south Atlantic occurs at both east and west coast locations in southern Africa. Backward trajectories originating at the west and east African coastal sites (Figure 5c and d) intersect some of the regions where fires are observed in southern Brazil. Trajectories from these sites do not originate over southern Africa.

The example from the low CO class shows a further decoupling of eastern southern Africa and the south Indian Ocean from the African subcontinent and from South America with extremely low concentrations of CO (< 75 ppbv) on the east coast of South Africa (Figure 3c). Figures 5 e and f show westerly transport extending well poleward to south of 50° S latitude with no trajectories originating in Brazil. Most of the air reaching the east coast of southern Africa originates south of 35° S. Air reaching the west coast site, however, shows evidence of ridging into southern Africa from the south Atlantic anticyclone, coupling with fires over southern Africa with westward transport across Africa in the easterlies occurring along about 16° S latitude and at altitudes below about 650 hPa.

4. Discussion and Conclusions

The fires shown in Figure 2 over South America and southern Africa lie mostly between the equator and 25° S. When the STAC is well established and shifted equatorward with a strong anticyclone over southern Africa, trajectory analyses at 450 hPa suggest that the biomass burning products being produced over the continent of South America and southern Africa can be transported between the continents across the Atlantic Ocean. High values of CO (> 150 ppbv) are seen in the atmospheric column by the MAPS instrument. High levels of CO are likely to be present at the altitude of the trajectories (450 hPa) based on the response of the instrument. Under strongly linked conditions, high values of CO also extend eastward across southern Africa to the central region of the southern Indian Ocean.

As the large scale circulation fields change, either by weakening the STAC over the south Atlantic or shifting the anticyclone toward the South Pole, coupling between the two continents ceases. Fires over both continents (Figure 2) during early October 1994 lie between the equator and 25° S. East-to-west transports shift poleward of the latitude of the fires and no longer carry high concentrations of CO from South America to southern Africa, or into the Indian Ocean.

In Case 3, low concentrations of CO are seen along the east coast of South Africa and in the western south Indian Ocean typical of Antarctic background air. High values of CO continue to be seen over western southern Africa, and over the south Atlantic where trajectories circulating anticyclonically can intercept and carry biomass burning products from the east. Under these circumstances (Case 3), South America is more likely to receive biomass burning products from southern Africa than the reverse, which may contribute to the high values of CO observed over Brazil.

These results suggest that significant intercontinental transport of biomass burning products can occur between South America and southern Africa. These transports are a function of large-scale circulation fields, particularly the strength and position of the large subtropical anticyclones.

The elevated mixing ratios of CO seen over the equatorial east Indian Ocean and extending southwestwards into the southern central Indian Ocean, cannot be directly ascribed to the observed fires over Australia (Figure 2). Transport analysis similar to that carried out over southern Africa would be required before the sources of this region of maximum CO could be inferred.

Biomass burning appears to be the dominant source of free tropospheric CO in the southern hemisphere as measured by the MAPS instrument during austral spring. Contributions from anthropogenic sources in the northern hemisphere are either concentrated in the lowest layers of the atmosphere and therefore not observed by MAPS, or near surface values are mixed and diluted through the column. Since the MAPS CO instrument reflects mainly the global distributions of CO at mid-tropospheric levels ($\sim$ 450 hPa) then at these levels biomass burning and not the anthropogenic sources is the main source of global CO during September-October.

The next generation of CO sensors, MOPITT on the EOS-AM platform, TES on EOS-PM and even MicroMAPS (currently not scheduled with a launch opportunity) would benefit greatly from a better understanding of dynamic meteorological processes that affect or redistribute CO, as well as other trace gases and particulates, in the troposphere. This increased understanding will lead to the development of improved calibration and validation programs for space-based sensors, to the identification of key factors that influence the global distribution of

these trace species, and the improvement of the capability of global models that seek to parameterize these dynamical and chemical processes.

Acknowledgements

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Figure Captions

Figure 1:   Zonally average CO mixing ratios in parts per billion volume

(ppbv) from April and October 1994 as seen by the MAPS instrument aboard the

Space Shuttle Endeavour (after Connors et al., 1998).

Figure 2:   Observations of fires and smoke by the astronauts aboard the

space shuttle Endeavor from 30 September to 11 October 1994.

Figure 3:   Tropospheric CO observed by MAPS on the space shuttle for

three cases during the period 30 September to 11 October 1994.

A)   High CO Case: 1 October 1994, CO > 150 ppbv, over South America,

southern Africa and the southern Atlantic and Indian Oceans.

B)   Moderate CO Case: 10 October 1994, 105 < CO < 150 ppbv, over

eastern southern Africa and the south Indian Ocean.

C)   Low CO Case: 7 October 1994, CO < 105 ppbv, over eastern southern

Africa and the south Indian Ocean.

Figure 4:   Geographic representation of location of the nine point grids

corresponding to the backward air parcel trajectory origins at 21°S 12°E and 25°S
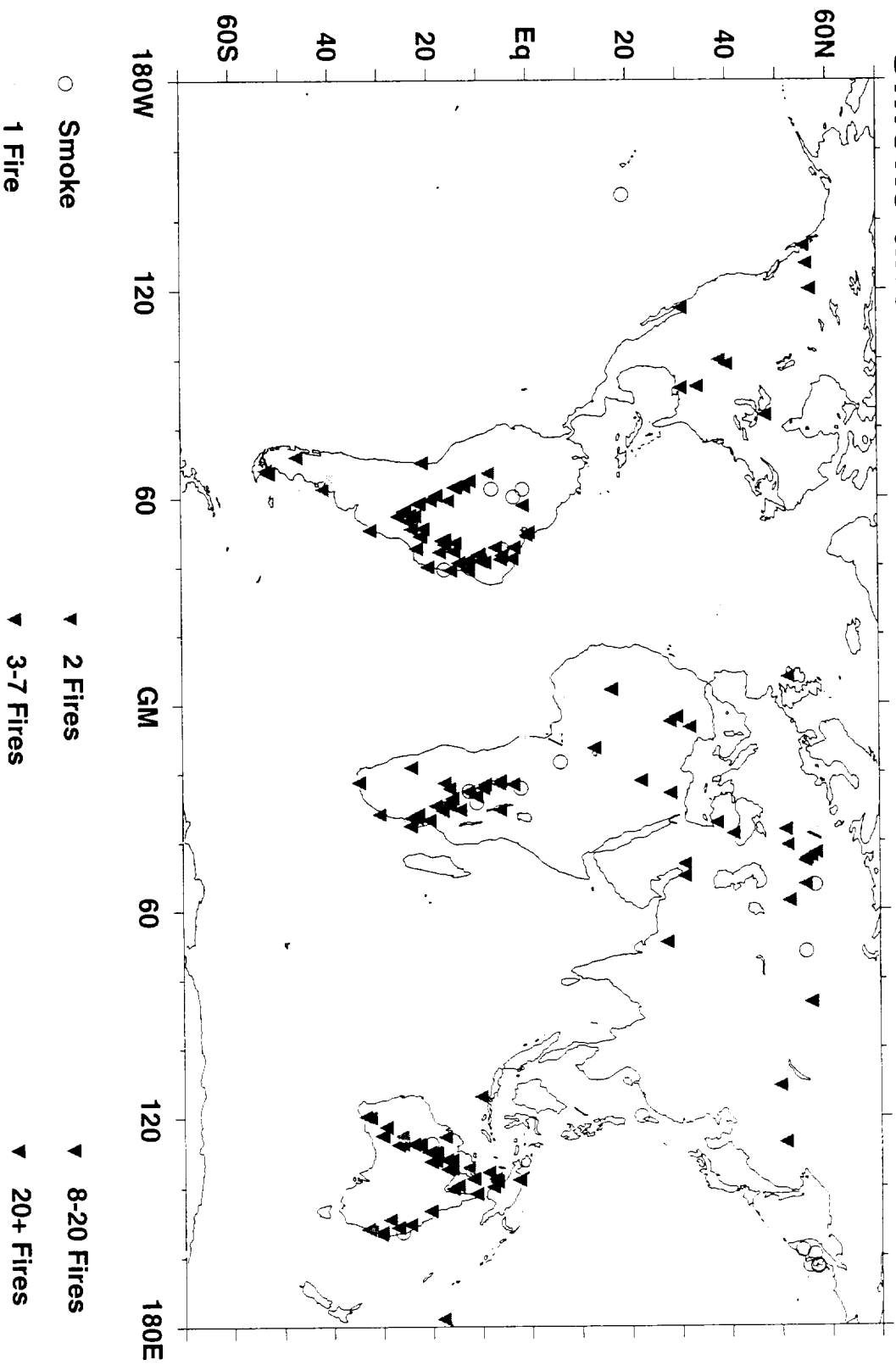
and 34°E, respectively.

Figure 5:   Ten-day backward trajectories from a 9 point 2° latitude x 2°

longitude grid located at 450 hPa for

a)   Case 1, 1 October 1994, with the west coast of southern Africa

(21° S, 12° E) as the air parcel trajectory origin.

b)   Case 1, 1 October 1994, with the east coast of southern

Africa (25° S, 34° E) as the air parcel trajectory origin.

c)   Case 2, 10 October 1994, with the west coast of southern

Africa (21° S, 12° E) as the air parcel trajectory origin.

d)   Case 2, 10 October 1994, with the east coast of southern

Africa (25° S, 34° E) as the air parcel trajectory origin.

e)   Case 3, 7 October 1994, with the west coast of southern

Africa (21° S, 12° E) as the air parcel trajectory origin.

f)   Case 3, 7 October 1994, with the east coast of southern

Africa (25° S, 34° E) as the air parcel trajectory origin.

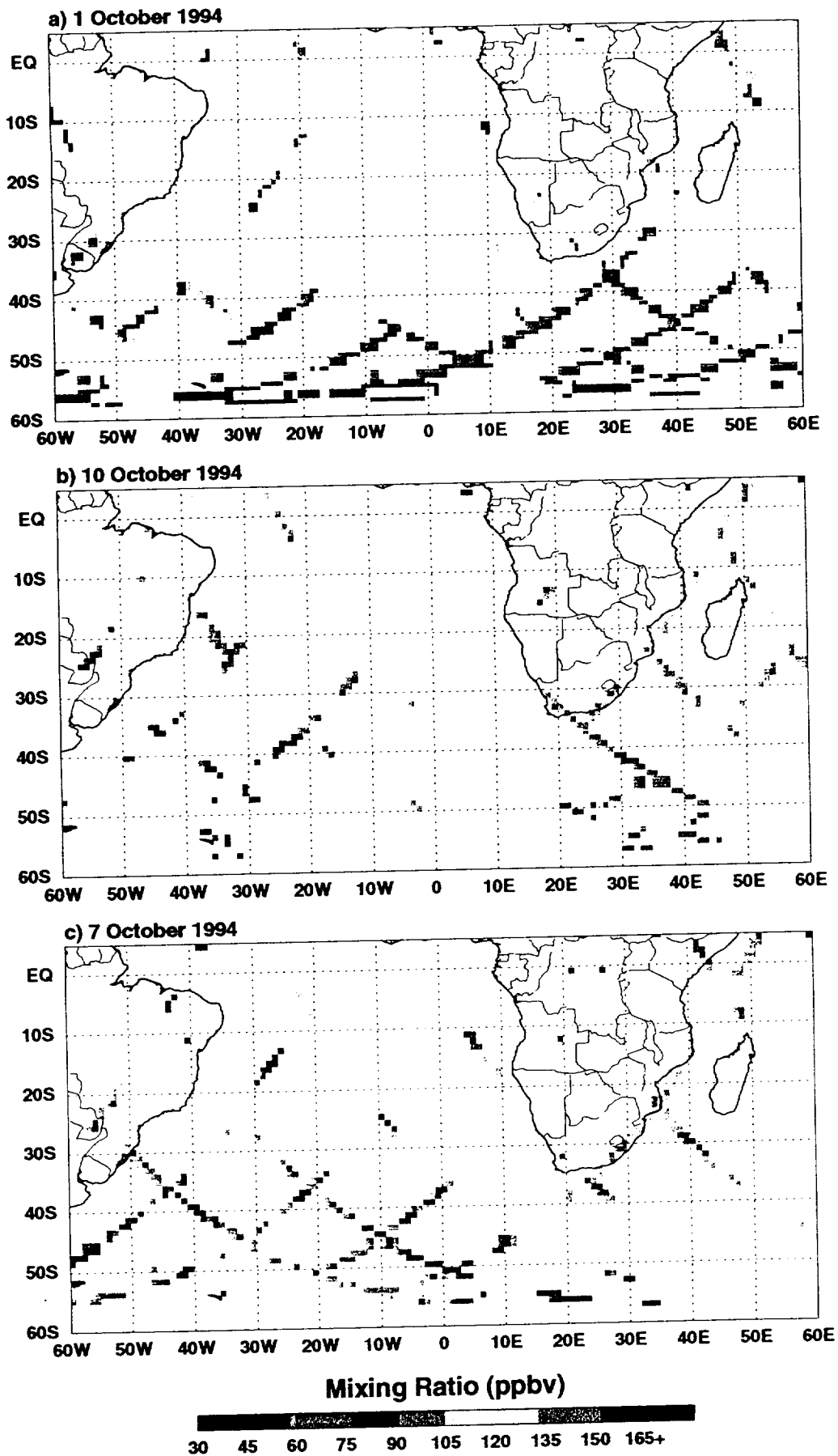Measurement of Air Pollution from Satellites
Tropospheric CO Zonal Averages

April 1994
October 1994

CO Mixing Ratio (ppbv)

Degrees Latitude
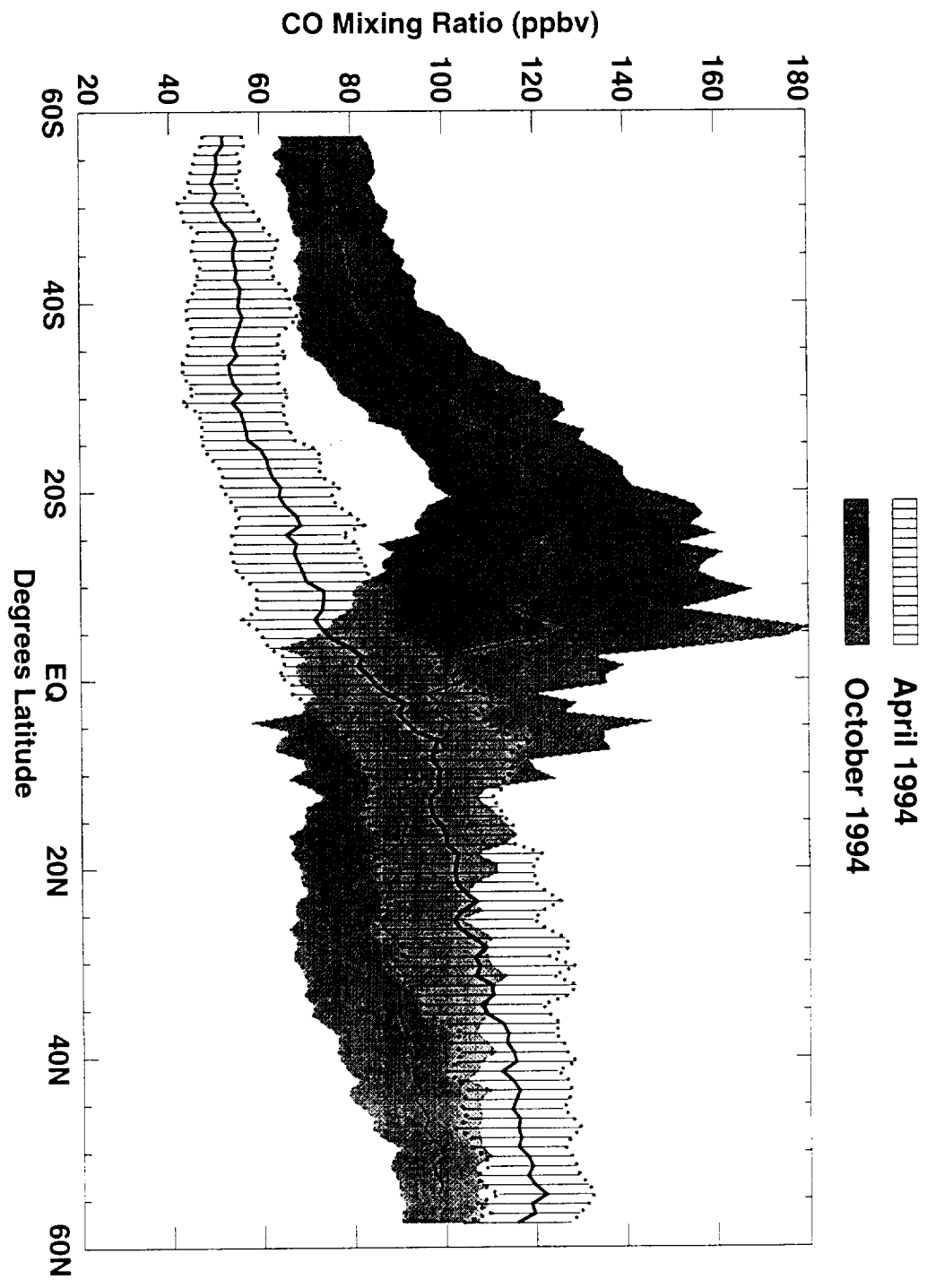
Smoke and Fires from STS-68 Astronaut Observations

SRL-2 September 30 - October 11, 1994

Smoke ○

1 Fire ▼

2 Fires ▼

3-7 Fires ▼

8-20 Fires ▼

20+ Fires ▼

MAPS Carbon Monoxide Measurements

a) 1 October 1994

b) 10 October 1994

c) 7 October 1994

Mixing Ratio (ppbv)

30 45 60 75 90 105 120 135 150 165+
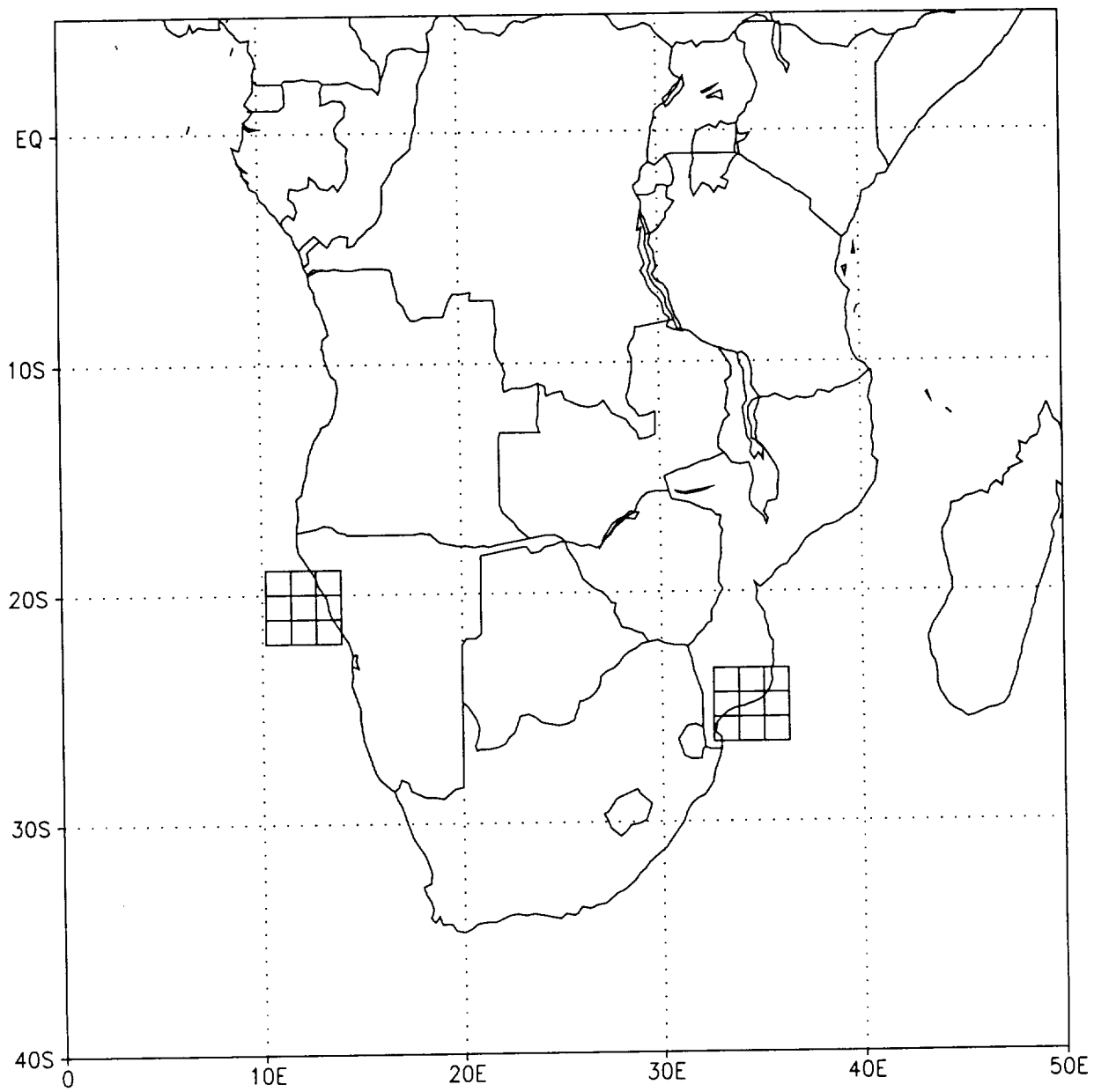
Figure 4

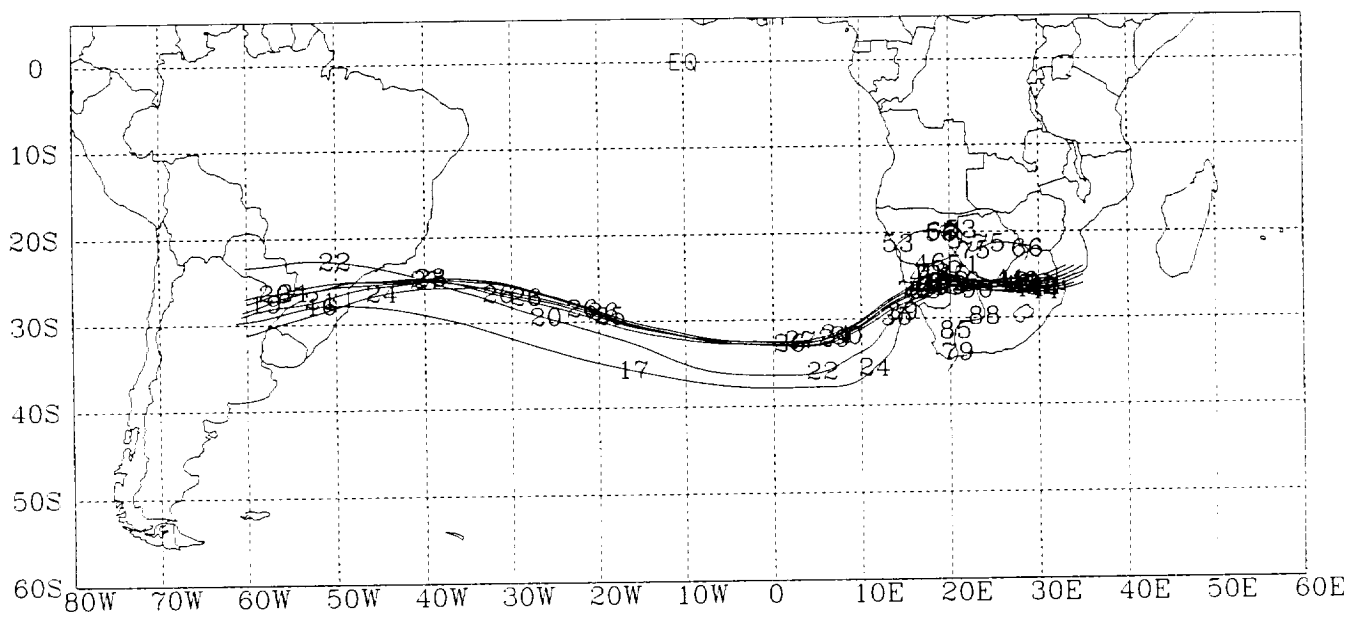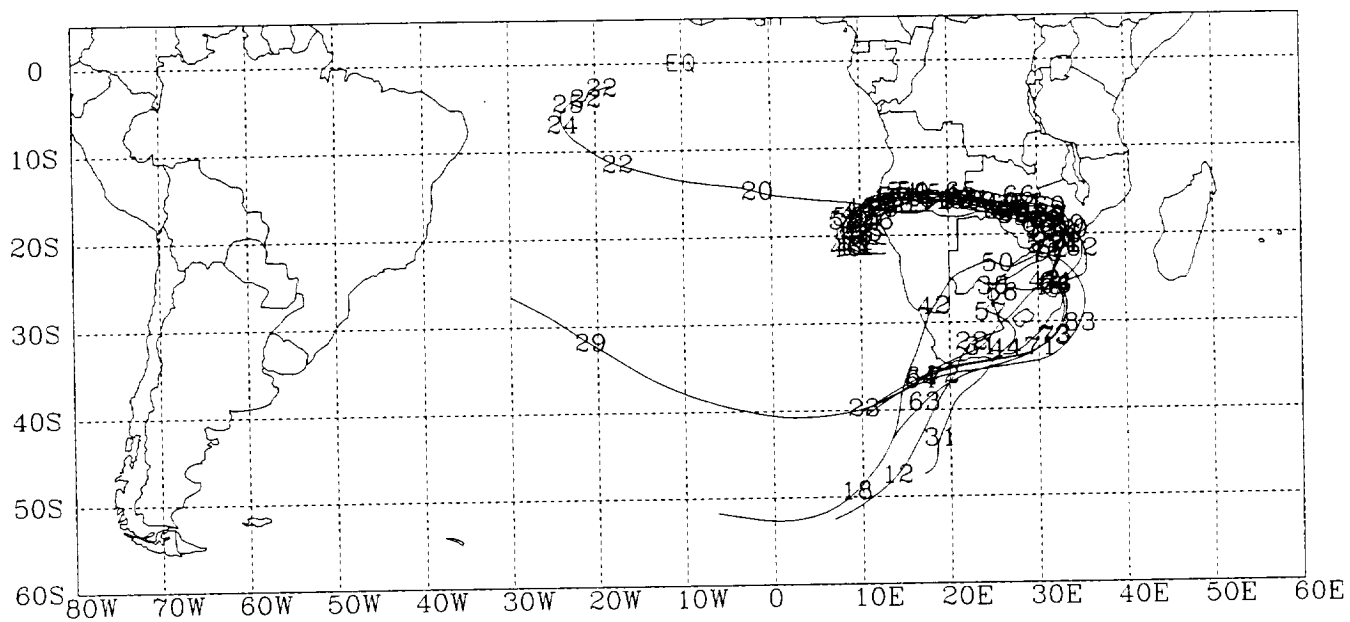Elevated CO Case

a.
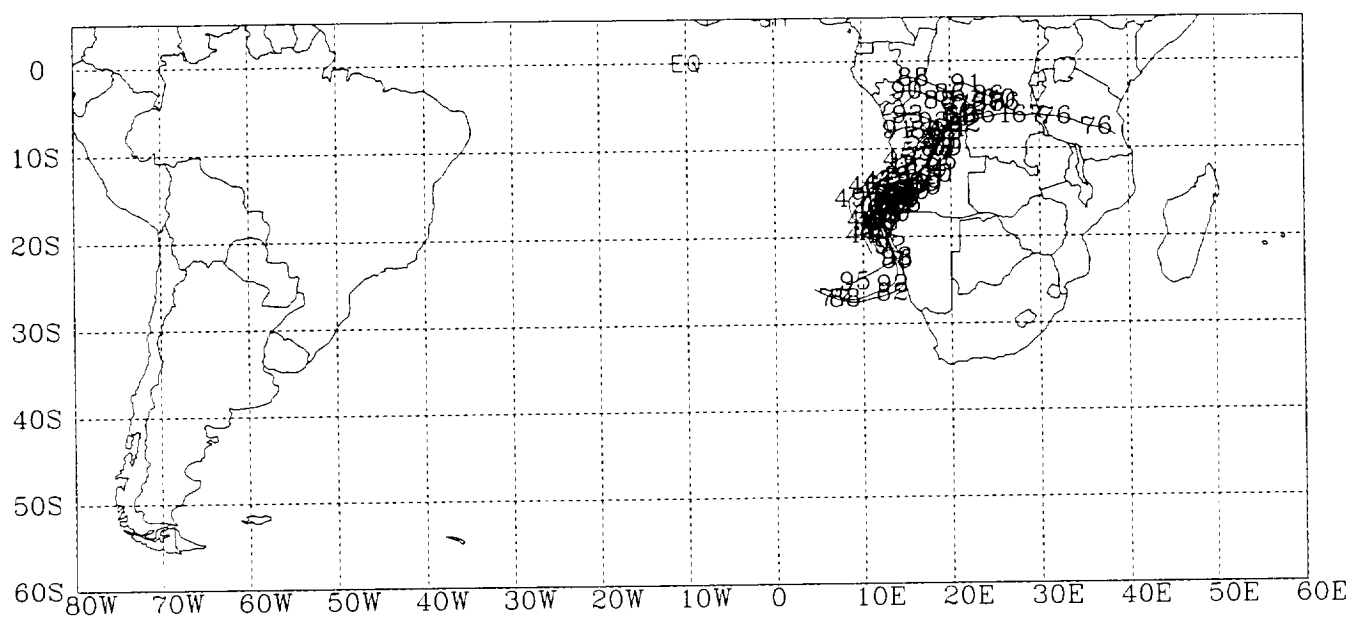
b.
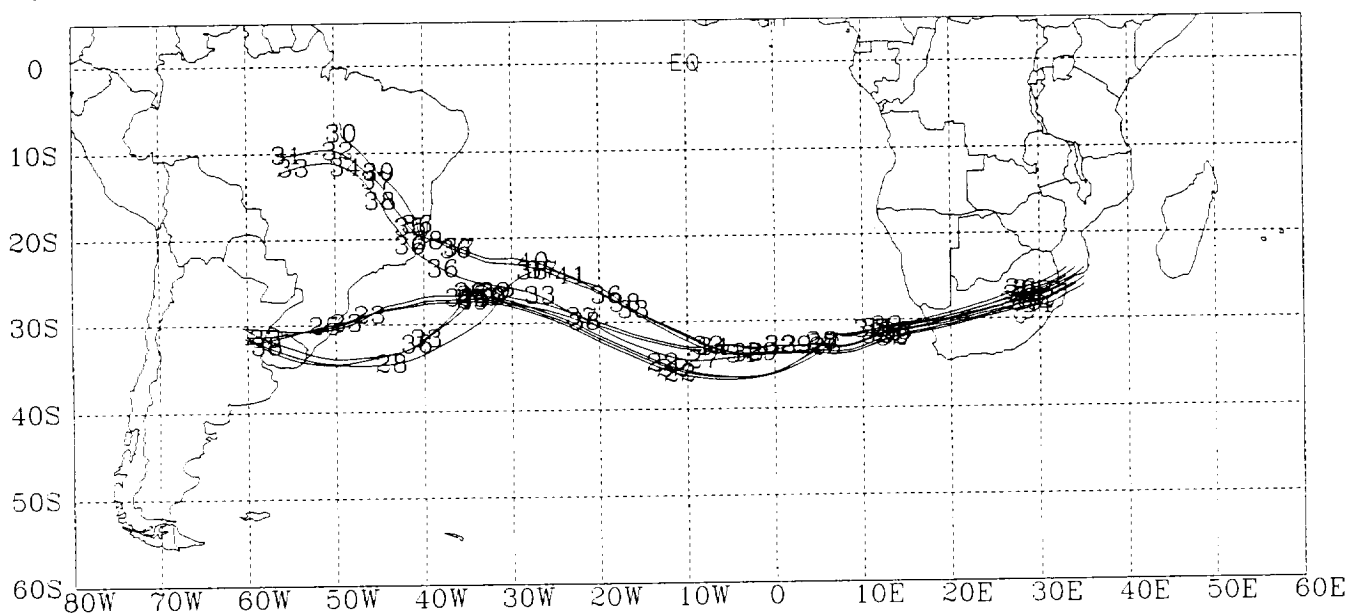
Figure 5
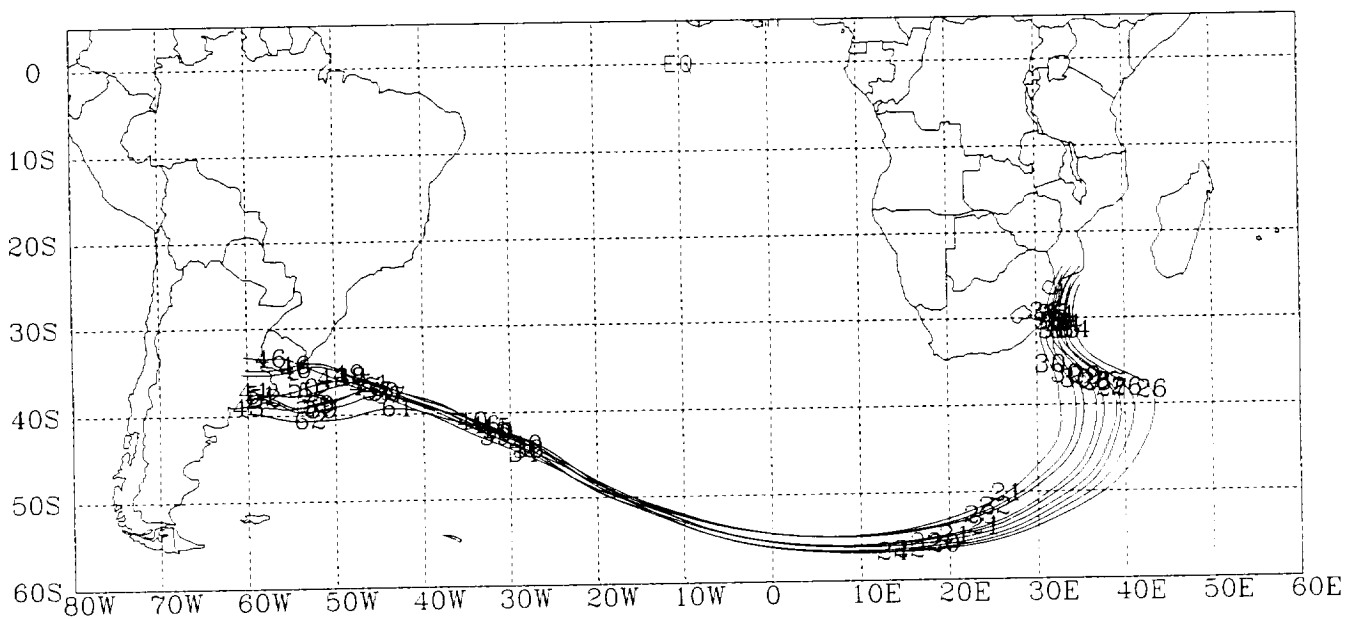
Moderate CO Case

c.
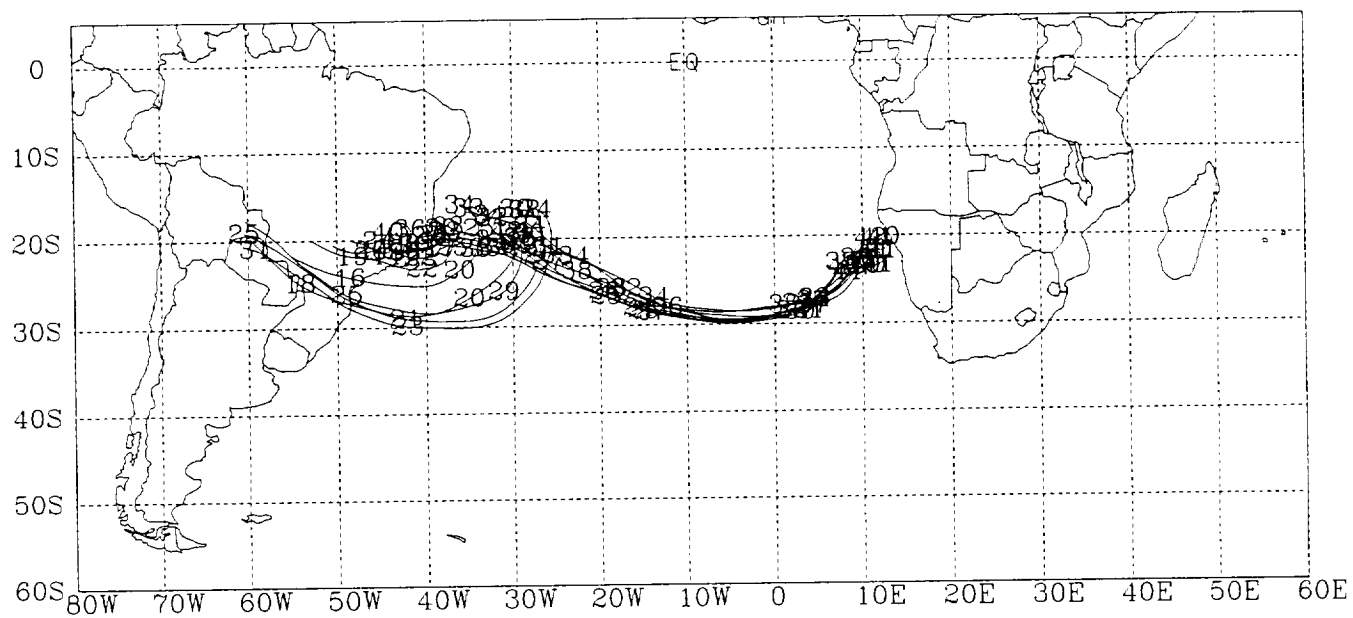
d.

Figure 5

Low CO Case

e.

f.

Figure 5